Non-local Quantum Electrodynamics. II. Possibility of correlated $2n$-photon absorption in gases leading to VERY High frequency spontaneous emission and Very high order Harmonic Generation

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ABSTRACT

In a recent work [1] we expounded a non-local Quantum Electrodynamics (QED) which predicted a *linear two-photon absorption* by an atom placed in a laser field of appropriate intensity and frequency. In this paper we extend our earlier work to show that the theory allows for linear $2n$-photon absorption by gaseous matter where, under suitable conditions, n may literally run upto thousands. The consequences of this extension of the theory are outlined and predictions are made which may be verified in laboratories.

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* this work is dedicated to his memory

I. INTRODUCTION

Recently [1] we showed that complex scalar fields remain covariant under a non-local Abelian gauge transformation, and showed how a non-local QED follows naturally from this gauge symmetry. (This paper will be referred to as I in the following.) The result can of course be trivially extended to spinor fields. From a practical point of view, the most important result of this nonlocal QED is that under suitable conditions that can be met in laboratory as well as cosmological environments, a *two-photon absorption linear in intensity* can occur from radiation fields travelling through gaseous matter. The impact of this new QED on atomic and molecular physics, particle physics and cosmology has been discussed in a series of papers [1-4]. In [2] we also suggested an experimental test for this abovementioned result.

In paper I, it has been shown [1] that the correlated two-photon absorption in atoms can give rise to probability which is linear in laser intensity. In this paper we will show that this correlated two-photon absorption process can be generalized to 2n-photon absorption process, where n can go upto thousands or more. Previously it has been shown that the correlated mode-pair of photons may exist under certain conditions, within the framework of non-local Quantum Electrodynamics (QED). By second quantization of this non-local field, correlated two-photon annihilation and creation operators were developed and it has been demonstrated that these operators operate on correlated photon number states by annihilating or creating two photons at a time respectively (one photon from each mode). Hence one can get correlated vacuum field in absence of any photons. Which means that the vacuum for the electromagnetic field is structured by the presence of this new type of non-local correlated zero-photon mode pairs. Here we will generalize this vacuum field by considering 2n correlated modes, where n can go upto a large number. But to make the zero point energy finite, one will have to put an upper limit to the value of n. The upper limit for the number n can be such that $n\hbar\omega \leq m_e c^2$ where m_e is the rest mass of electron, c is the velocity of light and ω is the frequency of the photon.

It has been shown before that the correlated two-photon absorption can occur in a single electron or two-electron (preferably correlated) system, under the conditions that (i) the time for two-photon absorption $\delta t << \frac{1}{\omega}$, where ω is the frequency of the photon and (ii) the number of photons (of energy flux I) passing through atomic volume V i.e. $\frac{VI}{c\hbar\omega}$ must be greater than or equal to the number of outer-shell electrons (n_e) in the atom. Of course, above number of photons should be present within the volume V during the time δt, which is much less than $\frac{1}{\omega}$, so that the absorption can occur during this time interval. Therefore, there are three parameters V, I and ω which can be controlled to satisfy the condition

$$\frac{VI}{c\hbar\omega^2} >> n_e \tag{1}$$

For a single atom, V is fixed and if ω is given, one will have to increase the laser intensity I, so that the number of photons exceeds the number of electrons present in the outer shell. On the other hand if V can be made large as in the case of cluster of atoms, the condition (1) can be easily satisfied for a lower laser intensity. In case of n-photon absorption, where n can be large on the order of 1000, the cluster will be a preferable candidate, since it can have radius one or two orders of magnitude larger than the single atom. Under this condition absorption of 1000s' of photons will be more efficient for clusters than for single atom for a given intensity of laser. Moreover, absorption of 1000s' of photons in a single atom, within the interval of time δt becomes a more stringent condition than that in cluster of atoms where a large number of electrons are available within the volume V.

In the previous paper it has been shown that the correlation among electrons may affect the phenomenon of correlated two-photon absorption by two-electron atoms. In this work we will not discuss about electron correlation, but it can be stated that the presence of electron correlation will also affect the $2n$ photon correlated absorption process.

Within the framework of non-local QED, the vacuum field is structured due to the presence of 2n correlated zero-photon modes and hence the electrons excited by 2n correlated photon absorption can spontaneuosly emit a single photon of energy $2n\hbar\omega_k$ due to the interaction with this correlated vacuum field. This is the phenomenon which can be attributed to the emission of x-rays observed [7] in clusters when irradiated with strong lasers of intensity $I \geq 10^{16} W/cm^2$.

Application of the above formalism in the area of matter-radiation interaction has so far been confined to one- and two-electron atoms. In the present paper it will be shown that the abovementioned property of nonlocal gauge transformation symmetry can be extended in such a manner as to be directly applicable to n-electron systems, where n can literally run upto thousands as for example in clusters. The results of the interaction of such systems with laser radiation can, according to this theory, be startling. One of the results that we are going to prove is the possibility of $2n$-photon absorption linear in intensity from a laser field, leading to the prospect of highly efficient $2n$-th order harmonic generation and/or spontaneous emission using gases consisting of n-electron atoms, molecules or clusters.

II. THEORY

We begin by giving a brief summary of the work presented in I.

For a wavefunction (or classical field) $\phi(x)$ where $x \equiv (\mathbf{r}, t)$, one of the ways to ensure the covariance (i.e. form-invariance) of the Lagrangian under an Abelian gauge transformation (GT) of the second kind $T = \exp(-ie\Lambda(x))$ [Λ being any arbitrary function of x] is to replace the ordinary partial derivative $\partial_\mu \equiv \partial/(\partial x^\mu)$ in the Lagrangian by a *covariant derivative* D_μ involving a dynamical field [5]. D_μ is defined by the property that under the above GT, $D_\mu\phi$ transforms in exactly the same manner as ϕ does ; i.e. for infinitesimal $\Lambda, \delta(D_\mu\phi) = -ie\Lambda D_\mu\phi$. For a charged particle one can define $D_\mu = \partial_\mu + ieA_\mu(x)$, where $A_\mu = (A_0, -\mathbf{A})$ is the electromagnetic field potential and e plays the role of the *coupling constant* of the latter with $\phi(x)$. To ensure covariance of D_μ under the above GT, the field potential A_μ should transform as $A_\mu \rightarrow A_\mu + (\partial_\mu\Lambda)$. This, of course, agrees with the *gauge arbitrariness* of A_μ allowed by the *gauge-independence* of the electromagnetic field tensor $F^{\mu\nu} = \partial^\mu A^\nu - \partial^\nu A^\mu$.

Against this elementary background, we introduced in [1] a *non-local* gauge transformation $\mathcal{T} = \exp(-ie\Lambda|x\rangle\langle x'|)$, whose action on ϕ is defined by

$$\mathcal{T}\phi \equiv \int \exp(-ie\Lambda(x, x'))\phi(x')\, d^4x' \qquad (1')$$

(A detailed discussion of this notation is given in [3], appendix A.) For a charged particle, this allows the construction of a *new covariant derivative*

$$\mathcal{D}_\mu = \frac{\partial}{\partial x^\mu} + \frac{\partial}{\partial x'^\mu} + ie\mathcal{A}_\mu|x\rangle\langle x'|$$

where the action of $\mathcal{D}_\mu$ on ϕ is to be understood in the sense of eqn. (1'), and where the nonlocal $\mathcal{A}$ transforms as $\mathcal{A}_\mu|x\rangle\langle x'| \to \mathcal{A}_\mu|x\rangle\langle x'| + \partial_\mu\Lambda|x\rangle\langle x'| + \partial'_\mu\Lambda|x\rangle\langle x'|$. The necessary and sufficient condition for gauge-independence of the corresponding nonlocal field tensor

$$\mathcal{F}^{\mu\nu}|x\rangle\langle x'| = (\partial^\mu + \partial'^\mu)\mathcal{A}^\nu|x\rangle\langle x'| - (\partial^\nu + \partial'^\nu)\mathcal{A}^\mu|x\rangle\langle x'| \tag{2}$$

is that the commutator $[\partial^\mu, \partial'^\nu]$ should vanish, i.e. the non-locality of the field should be of the EPR-type.

Under the action of this nonlocal field $\mathcal{A}$, a typical matrix element of an atomic transition $|i\rangle \to |f\rangle$ is given by

$$M_{fi}(t) = -\frac{e}{m}\langle\psi_f(x), n_f|\mathbf{p}.\mathcal{A}(x,x')|\psi_i(x'), n_i\rangle \tag{3}$$

the integration running over $d^3\mathbf{r}$ and d^4x'. We have used the Coulomb gauge.

A Fourier expansion of this nonlocal potential can be carried out (see, e.g. [6]) via a *double summation* over the usual photon modes :

$$\mathcal{A}(x,x') = \sum_{\mathbf{k}_1\lambda_1}\sum_{\mathbf{k}_2\lambda_2}[C_{\mathbf{k}\lambda}\hat{\epsilon}_{\mathbf{k}\lambda}(\hat{r},\hat{r}')\exp(i\mathbf{k}_1.\mathbf{r} - i\omega_{\mathbf{k}_1}t + i\mathbf{k}_2.\mathbf{r}' - i\omega_{\mathbf{k}_2}t') + c.c.] \tag{4}$$

Using standard methods (see any textbook on QED) the energy of this nonlocal field in a volume Ω is given by

$$W_2 = \frac{1}{2}\sum_{\mathbf{k}\lambda}(Y^2_{\mathbf{k}\lambda} + 4\omega^2_{\mathbf{k}}Z^2_{\mathbf{k}\lambda}) \tag{5}$$

where $Y_{\mathbf{k}\lambda} = -i\frac{\sqrt{\Omega/\pi}}{2c}2\omega_{\mathbf{k}}(C_{\mathbf{k}\lambda} - C^*_{\mathbf{k}\lambda}), Z_{\mathbf{k}\lambda} = \frac{\sqrt{\Omega/\pi}}{2c}(C_{\mathbf{k}\lambda} + C^*_{\mathbf{k}\lambda})$. Each mode subscript $\mathbf{k}\lambda$ here actually stands for a *phase-correlated mode pair* $(\mathbf{k}_1\lambda_1, \mathbf{k}_2\lambda_2)$, and $2\omega_{\mathbf{k}}$ stands for $\omega_{\mathbf{k}_1} + \omega_{\mathbf{k}_2}$. The quantities Y and Z satisfy Hamilton's canonical equations of motion with W_2 as the hamiltonian, and in I we quantized the nonlocal QED by requiring that Y and Z be q-numbers obeying the commutation relation

$$[Z_{\mathbf{k}\lambda}, Y_{\mathbf{k}'\lambda'}] = i\hbar\delta_{\mathbf{k}\mathbf{k}'}\delta_{\lambda\lambda'}.$$

Ultimately, the nonlocal field energy comes out in terms of a *new pair of creation and annihilation operators :*

$$W_2 = \frac{1}{2}\sum_{\mathbf{k}\lambda}\hbar\omega_{\mathbf{k}}(b_{\mathbf{k}\lambda}b^\dagger_{\mathbf{k}\lambda} + b^\dagger_{\mathbf{k}\lambda}b_{\mathbf{k}\lambda}), \quad \text{where} \quad (b, b^\dagger)_{\mathbf{k}\lambda} = \frac{1}{\sqrt{2\hbar\omega_{\mathbf{k}}}}(2\omega_{\mathbf{k}}Z_{\mathbf{k}\lambda} \pm iY_{\mathbf{k}\lambda})$$

Here b and $b^\dagger$ are *two-photon* annihilation and creation operators, obeying the new commutation relations :

$$[b_{\mathbf{k}\lambda}, b^\dagger_{\mathbf{k}'\lambda'}] = 2\delta_{\mathbf{k}\mathbf{k}'}\delta_{\lambda\lambda'}, \quad [b_{\mathbf{k}\lambda}, b_{\mathbf{k}'\lambda'}] = [b^\dagger_{\mathbf{k}\lambda}, b^\dagger_{\mathbf{k}'\lambda'}] = 0$$

A natural extension of the non-local GT (1) to the case of a two-particle system would be the transformation $\mathcal{T}_2|x_1,x_2\rangle\langle x_1', x_2'|$ which, written out in full, becomes

$$\mathcal{T}_2\phi(x_1,x_2) \equiv \int\int \exp(-ie\Lambda(x_1,x_2,x_1',x_2'))\phi(x_1',x_2')\,d^4x_1'\,d^4x_2' \tag{6}$$

This allows for a new covariant derivative

$$\mathcal{D}_{2\mu} = \partial_{1\mu} + \partial_{2\mu} + \partial'_{1\mu} + \partial'_{2\mu} + \mathcal{A}_{2\mu}|x_1,x_2\rangle\langle x_1',x_2'| \tag{7}$$

whereupon the matrix element for the transition $|i\rangle \to |f\rangle$ becomes

$$M_{2_{fi}}(t) = -\frac{e}{m}\langle\psi_f(x_1,x_2), n_f|(\mathbf{p}_1+\mathbf{p}_2).\mathcal{A}_2(x_1,x_2,x_1',x_2')|\psi_i(x_1',x_2'), n_i\rangle \tag{8}$$

Let us now extend this formalism to correlated 2n-photon absorption in a n-electron system, by using the fourier expansion [6] of non-local vector potential as follows:

$$\mathcal{A}(x_1,--,x_n,x_1',--,x_n') = \sum_{\mathbf{k}_1\lambda_1} -- \sum_{\mathbf{k}_n'\lambda_n'} [C_{\mathbf{k}\lambda}\hat{\epsilon}_{\mathbf{k}\lambda}(\hat{r}_1,--,\hat{r}_n,\hat{r}_1',--\hat{r}_n')$$

$$\exp(i\mathbf{k}_1.\mathbf{r}_1 + -- + i\mathbf{k}_n.\mathbf{r}_n - i\omega_{\mathbf{k}_1}t_1 - -i\omega_{\mathbf{k}_n}t_n)$$
$$X\exp(i\mathbf{k}_1'.\mathbf{r}_1' + -- + i\mathbf{k}_n'.\mathbf{r}_n' - i\omega'_{\mathbf{k}_1}t'_1 - -i\omega'_{\mathbf{k}_n}t'_n) + c.c] \tag{9}$$

Each mode subscript $\mathbf{k}\lambda$ here actually stands for n *phase-correlated mode pairs* $(\mathbf{k}_1\lambda_1, \mathbf{k}_1'\lambda_1{}'), --, (\mathbf{k}_n\lambda_n, \mathbf{k}_n'\lambda_n{}')$.

Hence the electric field amplitude $\mathcal{E}(x_1,--,x_n,x_1',--,x_n')$ can be obtained by evaluating the time derivatives of A with respect to times $t_1,--t_n'$. By using the usual method one can obtain the field energy density in terms of new c-numbers $Y_{N\mathbf{k}\lambda}$ and $Z_{N\mathbf{k}\lambda}$ where

$$Y_{N\mathbf{k}\lambda} = -i\frac{\sqrt{\Omega/\pi}}{2c}N\omega_{\mathbf{k}}(C_{\mathbf{k}\lambda} - C^*_{\mathbf{k}\lambda})$$

,

$$Z_{N\mathbf{k}\lambda} = \frac{\sqrt{\Omega/\pi}}{2c}(C_{\mathbf{k}\lambda} + C^*_{\mathbf{k}\lambda})$$

. Here $N\omega_{\mathbf{k}}$ stands for $\omega_{\mathbf{k}_1} + -- + \omega'_{\mathbf{k}_n}$. The quantities Y and Z satisfy Hamilton's canonical equations of motion with the total field energy as the hamiltonian. To second quantize the nonlocal QED, Y and Z are considered to be q-numbers obeying the commutation relation

$$[Z_{N\mathbf{k}\lambda}, Y_{N\mathbf{k}'\lambda'}] = i\hbar\delta_{\mathbf{k}\mathbf{k}'}\delta_{\lambda\lambda'}.$$

Ultimately, the field energy can be written in terms of a new pair of *creation and annihilation operators* defined as

$$(b_N, b_N^\dagger)_{\mathbf{k}\lambda} = \frac{1}{\sqrt{2\hbar\omega_k}} N\omega_{\mathbf{k}}(Z_{N\mathbf{k}\lambda} \pm iY_{N\mathbf{k}\lambda}) \tag{10}$$

these operators obey the commutation relations as follows

$$[(b_N)_{\mathbf{k}\lambda}, (b^\dagger{}_N)_{\mathbf{k}'\lambda'}] = N\delta_{\mathbf{k}\mathbf{k}'}\delta_{\lambda\lambda'}, \quad [(b_N)_{\mathbf{k}\lambda}, (b_N)_{\mathbf{k}'\lambda'}] = [(b_N^\dagger)_{\mathbf{k}\lambda}, (b_N^\dagger)_{\mathbf{k}'\lambda'}] = 0$$

These annihilation and creation operators operate on the correlated phopton number states as annihilating N number of photons or creating N muber of photons respectively as follows:

$$(b_N)_{\mathbf{k}\lambda}|n_{\mathbf{k}\lambda}\rangle = \sqrt{n}_{\mathbf{k}\lambda}|n_{\mathbf{k}\lambda} - N\rangle$$

and

$$(b_N^\dagger)_{\mathbf{k}\lambda}|n_{\mathbf{k}\lambda}\rangle = \sqrt{n_{\mathbf{k}_\lambda} + N}|n_{\mathbf{k}\lambda} + N\rangle \tag{11}$$

Therefore $(b_N)_{\mathbf{k}\lambda}$ and $(b^\dagger{}_N)_{\mathbf{k}\lambda}$ are the N-photon annihilation and creation operators respectively. Hence the vector potential and the field amplitude can be expressed in terms of these operators as shown in paper I. Therefore the dipole transition matrix elements for correlated 2n-photon absorption by n electrons (such as in clusters) can be given as:

$$\begin{aligned}\mathcal{X}_{Nfi} = \frac{\sqrt{2\pi I}}{c}\langle\psi_{l_f},(\mathbf{r}_1, -- \mathbf{r}_n)|(\mathbf{r}_1 + ---- + \mathbf{r}_n)\\ .\hat{\epsilon}_{N\mathbf{k}\lambda}(\hat{r}_1, ----, \hat{r}'_n)|\psi_{l_i}(\mathbf{r}'_1, -- \mathbf{r}'_n)\end{aligned} \tag{12}$$

where the polarization vector $\hat{\epsilon}_{N\mathbf{k}\lambda}$ can be expressed as

$$\hat{\epsilon}_{\mathbf{k}\lambda}(\hat{r}_1, --, \hat{r}_n, \hat{r}'_1, --, \hat{r}'_n) = \sum_j a_j \Pi_i(\hat{r}_i + \hat{r}'_i) P_j(\hat{r}_i.\hat{r}'_i)$$

where j varies from 0 to ∞ and $i = 1, 2, ---n$ Mod square of the matrix element $\mathcal{X}_{Nfi}$ will give rise to N-photon absorption probability which varies linearly with laser intensity. One can get the selection rule for the angular momentum by considering the angular integrals (as shown in paper I) as $l_f = l_i$ and $l_f = l_i + N$

III. DISCUSSIONS

It has been shown before, that the non-local theory has wide applications [1-4] in the fields of atomic/molecular physics, cosmology and particle physics. Recently it has been applied to explain the phenomenon of direct double ionization in atoms [8] in intense laser fields. Here it has been shown that the non-local model of QED can also be applied to explain the emission of x-rays from cluster of atoms when irradiated with strong laser field of intensity $> 10^{16} W/cm^2$. In cluster of atoms the radius of the cluster can be 10 times larger than that of a single atom and hence the volume can be 10^3 times larger than that of a single atom. It is obvious that the intensity (I) for correlated 2n-photon excitation can be atleast three orders of magnitude less than that required for the single atom (consider the relation (1)), in particular for a large number (can be 1000 or more) of photon absorption. Therefore the efficiency of emission will increase with the increase in the size of the clusters. But there is a limit in the increase of the size of clusters, since for efficient absorption the condition (1) should be satisfied. Increase in the size of the cluster means addition of more atoms and hence increase in the available number of electrons (n_e). Therefore if the increase in n_e is faster than the increase in V, the condition (1) fails to be satisfied. Moreover emission from clusters of high-Z atoms will be more effective, since in that case the available number of electrons will be more for the same size of the the ciuster of low-Z atoms. Furthermore, there exists a cutoff intensity below which x-ray emission from a cluster cannot be obtained (see quation (1)). For x-ray emission in a particular region of wavelength, the cutoff intensity will be different for clusters of different atoms. According to this theory, since the 2n-photon correlated absorption should occur during a very short time $<< \frac{1}{\omega}$, this phenomenon will be more effective for short-pulsed lasers. Some of the above mentioned features have already been observed in recent experiments [7] on x-ray emission from clusters of different atoms in presence of strong laser fields. Moreover in recent experiments [7] it has been found that together with the sharp higher order harmonic generations, x-ray emissions of relatively broad spectrum occur and persist for a long time. It has been shown above that once the atoms in cluster are excited by the absorption of correlated 2n-photons, can emit spontaneously (photons of energy around $2n\hbar\omega$) due to the interaction of the excited atom/highly charged ions with the vacuum of correlated modes. Since the duration of spontaneous emission depends on the natural linewidth of the states from which it is emitted, it can be as long as few nanoseconds [7]. Let us consider a situation under which spontaneous emission of duration 1 nanosecond can occur from a highly excited atom/highly charged ion. If the duration of spontaneous emission of $12ev$ photon can be of the order of 100 ns (e.g. spontaneous emission from $B^1\Sigma_u$ state to the ground state of H_2 molecule), then for $1200ev$ photon the duration of spontaneous emission can be of the order of 1 nanosecond, if the dipole transition moment from the ground state of the atom to the highly excited state is less in magnitude by two orders from that of H_2 molecules (between the first excited state and the ground state). Hence it is likely that the spontaneous emission from clusters can persist for 1 or two nanoseconds. Another possibility is that the innershell ionization of atoms in clusters can occur due to the highly efficient correlated 2n-photon absorption, followed by x-ray emission.

The wavelength dependence of x-ray emission from cluster has been observed to be more efficient for shorter wavelength lasers (248 nm) than that for the longer one (800 nm). This may be due to the fact that for long wavelength lasers the number of photons required for a particular transition is much larger than that for short wavelength lasers. Therefore it will lead to higher order transitions and hence correlation may be destroyed during the process of absorption.

Most striking feature of this type of absorption in the non-local picture of the electromagnetic field is that in this correlated 2n-photon absorption process, the probability for absorption varies linearly with laser intensity, thus making the absorption process much more efficient than that for multiphoton transition in the local picture of the electromagnetic field. Therefore within the framwework of non-local QED, the 2n-photon correlated absorption and hence the spontaneous emission/ higher order harmonic generation from the cluster will be very much effective in presence of very strong laser ($I > 10^{16} W/cm^2$) field.

Moreover in the field of interaction of matter with radiation, one can predict that if the 2n-photon correlated absorption of radiation by the electrons in matter is feasible (in this model of non-local QED), leading to processes such as pair production, the phenomenon of pair production will be visible for much lower intensity than that required for conventional transitions.

In conclusion, it has been shown that the two-photon correlated absoption model in the non-local QED can be extended to any arbitrary 2n-photon correlated absorption and hence leading to efficient harmonic generation and/or spontaneous emission due to the interaction with structured vacuum (of correlated modes). This type of absorption can occur in cluster of atoms when irradiated with short-pulsed intense radiation field and the efficiency of this process will be much greater than the conventional multiphoton processes, since the 2n-photon absorption in the non-local model is linearly dependent on laser intensity. With this model of non-local QED, some experimentally observed features could be explained and some predictions have been made which can be verified in the laboratory.

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